

PATENT ABSTRACTS OF JAPAN

(11)Publication number : 10-214682

(43)Date of publication of application : 11.08.1998

(51)Int.Cl.

H05B 33/00

C23C 14/56

H05B 33/10

(21)Application number : 09-016564

(71)Applicant : MITSUBISHI CHEM CORP

(22)Date of filing : 30.01.1997

(72)Inventor : TANAMURA MITSURU

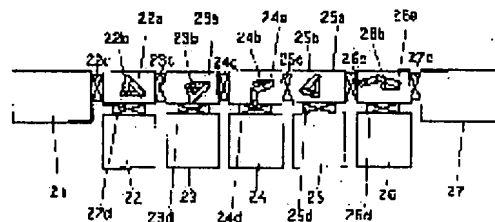
SATO YOSHIHARU

(54) MANUFACTURING DEVICE AND MANUFACTURE OF ORGANIC ELECTROLUMINESCENT ELEMENT

(57)Abstract:

PROBLEM TO BE SOLVED: To prevent respective layer-shaped deposits from contacting with moisture and oxygen in the atmosphere, and inexpensively manufacture an organic electroluminescence element excellent in a light emitting service life characteristic by consistently processing a base board without being exposed to the atmosphere in respective processes and a transfer process up to sealing after a pattern of an anode is formed.

SOLUTION: A base board on which an anode is formed as a pattern is arranged in a dry box 21, and after a base board surface is cleaned, it is sufficiently substituted with inert gas. Afterwards, the base board is carried and processed in order in working vacuum chambers 22 to 26 by respective prescribed operations of gate valves 22c to 27c and 22d to 26d, carrying vacuum chambers 22a to 26a and base board and mask carrying robot arms 22b to 26b. After respective layers are formed on the base board by these processings, the base board is not exposed to the atmosphere until a seal is completed by the dry box 27 filled with inert gas. Therefore, an organic electroluminescence element excellent in a light emitting characteristic and a light emitting service life characteristic can be inexpensively manufactured with



excellent productivity.

LEGAL STATUS

[Date of request for examination] 15.04.2002

[Date of sending the examiner's decision of rejection] 05.04.2005

[Kind of final disposal of application other than the examiner's decision of rejection or application converted registration]

[Date of final disposal for application]

[Patent number]

[Date of registration]

[Number of appeal against examiner's decision of rejection]

[Date of requesting appeal against examiner's decision of rejection]

[Date of extinction of right]

*** NOTICES ***

JPO and INPIT are not responsible for any damages caused by the use of this translation.

1. This document has been translated by computer. So the translation may not reflect the original precisely.
2. **** shows the word which can not be translated.
3. In the drawings, any words are not translated.

CLAIMS

[Claim(s)]

[Claim 1] In the equipment for manufacturing the organic electroluminescence devices which come to prepare two or more stratified deposits on a substrate The vacuum chamber for conveyance with carrying-in opening and taking-out opening of a substrate, and the operating vacuum chamber which stands in a row through the bulb which can pass [that vacuum transfer cutoff is possible and] said substrate in this vacuum chamber for conveyance, A migration means to be installed in this vacuum chamber for conveyance, and to transport a substrate into this operating vacuum chamber from said carrying-in opening, and to transport to said taking-out opening from this operating vacuum chamber, It is the organic electroluminescence-devices manufacturing installation equipped with the 1st thru/or the n-th unit the migration which has this processing means for stratified deposit formation established in this operating vacuum chamber, and for processing. So that the substrate introduced in this manufacturing installation may be transported from carrying-in opening of the 1st unit even to the n-th unit through this unit The manufacturing installation of the organic electroluminescence devices characterized by connecting taking-out opening of the unit of the substrate migration direction upstream to carrying-in opening of the unit of the substrate migration direction downstream.

[Claim 2] The manufacturing installation of the organic electroluminescence devices which are equipment for manufacturing the organic electroluminescence devices which come to prepare the seal member surrounding this stratified deposit while preparing two or more stratified deposits on a substrate, and are characterized by to have the facility for preparing said seal member, without exposing to atmospheric air after forming all stratified deposits in the manufacturing installation of the organic electroluminescence devices which prepared the following stratified deposit, without exposing to atmospheric air after preparing one stratified deposit.

[Claim 3] The manufacturing installation of the organic electroluminescence devices characterized by the equipment for preparing said stratified deposit on a substrate being a manufacturing installation according to claim 1 in claim 2.

[Claim 4] The manufacture approach of the organic electroluminescence devices characterized by to prepare said seal member, without exposing to atmospheric air after forming in the approach of manufacturing the organic electroluminescence devices which come to have the stratified deposit by which deposition formation was carried out, and which has an anode plate, an organic luminous layer, and cathode at least, and a seal member surrounding this stratified deposit to the shape of a layer on a substrate and this substrate, without exposing an organic luminous layer and cathode to atmospheric air within a vacuum tub at least among these stratified deposits.

[Translation done.]

*** NOTICES ***

JPO and INPIT are not responsible for any damages caused by the use of this translation.

1. This document has been translated by computer. So the translation may not reflect the original precisely.
2. **** shows the word which can not be translated.
3. In the drawings, any words are not translated.

DETAILED DESCRIPTION

[Detailed Description of the Invention]

[0001]

[Field of the Invention] This invention is the equipment and the approach of manufacturing the thin film mold light emitting device which emits light, applying electric field to the luminous layer which consists of an organic compound in detail about the manufacturing installation and its manufacture approach of organic electroluminescence devices, and relates to the equipment and the approach of mass production nature being good and manufacturing efficiently organic electroluminescence devices excellent in the luminescence life property by low cost.

[0002]

[Description of the Prior Art] Organic electroluminescence devices have the descriptions, such as spontaneous light, a thin shape, and a high angle of visibility, and attract attention as the new source of flat-surface mold luminescence thru/or a display device.

[0003] In order to manufacture organic electroluminescence devices conventionally After forming transperence electric conduction film, such as ITO (indium stannic-acid ghost), by approaches, such as a spatter, on a glass substrate, After carrying out pattern processing, forming a lower electrode, installing this substrate in a vacuum deposition tub and forming an organic luminous layer etc. by approaches, such as heating vacuum evaporatio, It once took out from this vacuum tub, and the metal layer, the protective layer, etc. were formed by another vacuum tub, it took out from this vacuum tub after that, and sealing of the component section was performed.

[0004] By the above-mentioned conventional approach, in order to form multilayer structure, such as an organic electron hole transporting bed, an organic luminous layer, cathode, and a protective layer, the vacuum tub was returned to ordinary pressure from the vacuum each time, the raw material needed to be replaced and masks needed to be exchanged. Moreover, even when two or more vacuum tubs were prepared, the substrate between vacuum tubs needed to be changed. For this reason, since a substrate was exposed to atmospheric air between previous vacuum evaporatio and the next vacuum evaporatio and a vacuum evaporatio layer was exposed to oxygen and the moisture in atmospheric air, there was a problem that the property of a component fell. Moreover, in order to repeat the vacuum suction of a vacuum tub, and ordinary pressure release, production time was long and the trouble of being bad also had productive efficiency.

[0005] The approach of delivering a substrate and carrying out sequential vacuum evaporatio, having arranged two or more vacuum deposition tubs, and maintaining a vacua at the surroundings of it for the meantime focusing on one vacuum conveyance tub, for the purpose of performing the vacuum deposition process from after pattern processing of the transperence electric conduction film to formation of a protective layer consistently as an approach of solving these troubles, is tried (JP,8-111285,A).

[0006]

[Problem(s) to be Solved by the Invention] However, by this approach, since; substrate which does not turn to the mass production machine with which a conveyance tub will grow large especially if;

substrate size which has a limitation in the number of the vacuum evaporations which can be arranged around a conveyance tub becomes large from a structural feature will be discharged outside from a vacuum tub in front of a seal process after the protection stratification and will be exposed to atmospheric air, it had troubles, such as; with the difficult reinforcement of a component.

[0007] this invention person solves the above-mentioned conventional trouble, and it aims at offering the equipment and the approach of mass production nature being good and manufacturing the organic electroluminescence devices which were excellent in the property by low cost.

[0008]

[Means for Solving the Problem] In equipment for the manufacturing installation of the organic electroluminescence devices of claim 1 to manufacture the organic electroluminescence devices which come to prepare two or more stratified deposits on a substrate The vacuum chamber for conveyance with carrying-in opening and taking-out opening of a substrate, and the operating vacuum chamber which stands in a row through the bulb which can pass [that vacuum transfer cutoff is possible and] said substrate in this vacuum chamber for conveyance, A migration means to be installed in this vacuum chamber for conveyance, and to transport a substrate into this operating vacuum chamber from said carrying-in opening, and to transport to said taking-out opening from this operating vacuum chamber, It is the organic electroluminescence-devices manufacturing installation equipped with the 1st thru/or the n-th unit the migration which has this processing means for stratified deposit formation established in this operating vacuum chamber, and for processing. It is characterized by connecting taking-out opening of the unit of the substrate migration direction upstream to carrying-in opening of the unit of the substrate migration direction downstream so that the substrate introduced in this manufacturing installation may be transported from carrying-in opening of the 1st unit even to the n-th unit through this unit.

[0009] If it is the manufacturing installation of these organic electroluminescence devices, the vacuum chamber for conveyance of the 1st unit, The operating vacuum chamber of the 1st unit, the vacuum chamber for conveyance of the 1st unit, the vacuum chamber for conveyance of the 2nd unit, The operating vacuum chamber of the 2nd unit, and the vacuum chamber for conveyance of the 2nd unit, by carrying out sequential migration of the substrate in the order of the vacuum chamber for conveyance of the unit of ** (n-1), the vacuum chamber for conveyance of the n-th unit, the operating vacuum chamber of the n-th unit, and the vacuum chamber for conveyance of the n-th unit Without exposing a substrate to atmospheric air, a vacuum can be maintained and a stratified deposit can be formed efficiently (in addition). It is also possible to form a stratified deposit, without going via the operating vacuum chamber of the unit of arbitration, or it is also possible to return from the 3rd unit to the 2nd unit, and to form a stratified deposit.

[0010] And since the unit this migration and for processing is connected by the vacuum chamber for conveyance, the number of arbitration can be connected and the number of operating vacuum chambers for forming a stratified deposit is not restricted. It can respond also to the increment in the stratified deposit which duplication of this unit is also easy a deposit and vapor-deposits it easily, and a manufacturing cost can be held down. Moreover, even when substrate size is large, the vacuum chamber for conveyance does not grow large.

[0011] The manufacturing installation of the organic electroluminescence devices of claim 2 is equipment for manufacturing the organic electroluminescence devices which come to prepare the seal member which surrounds this stratified deposit while preparing two or more stratified deposits on a substrate. After forming all stratified deposits in the manufacturing installation of the organic electroluminescence devices which prepared the following stratified deposit, without exposing to atmospheric air after preparing one stratified deposit, it is characterized by having the facility for preparing said seal member, without exposing to atmospheric air.

[0012] If it is the manufacturing installation of these organic electroluminescence devices, after forming a stratified deposit in a substrate, the seal member surrounding this stratified deposit can be prepared without exposing a substrate to atmospheric air, and organic electroluminescence devices excellent in the luminescence life property can be manufactured.

[0013] As for the equipment for preparing a stratified deposit on a substrate, in this equipment, it is desirable that it is said manufacturing installation according to claim 1.

[0014] The stratified deposit with which deposition formation of the manufacture approach of the organic electroluminescence devices of claim 4 was carried out on the substrate and this substrate at the shape of a layer and which has an anode plate, an organic luminous layer, and cathode at least, In the approach of manufacturing the organic electroluminescence devices which come to have a seal member surrounding this stratified deposit, after forming without exposing an organic luminous layer and cathode to atmospheric air within a vacuum tub at least among these stratified deposits, it is characterized by preparing said seal member, without exposing to atmospheric air.

[0015] By this approach, since it carries out without exposing a substrate to atmospheric air consistently from formation of a stratified deposit to formation of a seal member, the organic electroluminescence devices which were remarkably excellent in the luminescence property and the luminescence life property can be manufactured.

[0016]

[Embodiment of the Invention] With reference to a drawing, the gestalt of operation of this invention is explained below at a detail.

[0017] First, the configuration of the organic electroluminescence devices manufactured by this invention with reference to drawing 2 -5 is explained.

[0018] Drawing 2 -4 are the typical sectional view showing the example of structure of the organic electroluminescence-devices body of the organic electroluminescence devices manufactured by this invention, and drawing 5 is the typical sectional view showing the organic electroluminescence devices which come to carry out sealing of such an organic electroluminescence-devices body by the seal member.

[0019] drawing 2 -5 -- setting -- 1 -- a substrate and 2 -- an anode plate and 3 -- an organic luminous layer and 3a -- an electron hole transporting bed and 3b -- for cathode and 5, a protective layer and 6 are [an electron transport layer and 3c / a hole injection layer and 4 / tooth-back glass, and 10, 10A and 10B of a sealing compound and 7] organic electroluminescence-devices bodies.

[0020] A substrate 1 serves as a base material of organic electroluminescence devices, and to excel in properties, such as optical property, thermal-resistance, surface precision, mechanical-strength, lightweight nature, and gas barrier property, is demanded. As a substrate 1, generally, although the plate of a quartz or glass, a metal plate, a metallic foil and a plastic film, a sheet, etc. are used, transparent synthetic-resin substrates, such as a glass plate, and polyester, the poly meta acrylic rate, a polycarbonate, the poly ape phone, are suitable.

[0021] The anode plate 2 formed on the substrate 1 plays the role of the hole injection to the organic luminous layer 3. As for this anode plate 2, conductive polymers, such as halogenation metals, such as metallic oxides, such as oxide of metals, such as aluminum, gold, silver, platinum, nickel, palladium, and platinum, an indium, and/or tin, and copper iodide, carbon black or Pori (3-methylthiophene), polypyrrole, and the poly aniline, etc. are usually preferably formed of an indium stannic-acid ghost (ITO). Formation of an anode plate 2 is usually performed by the sputtering method, a vacuum deposition method, etc. in many cases. When using particles, such as metal particles, such as silver, and copper iodide, carbon black, a conductive metallic-oxide particle, conductive polymer impalpable powder, etc., this can be distributed in a suitable binder resin solution, and an anode plate 2 can also be formed by applying on a substrate 1. Moreover, when using a conductive polymer, an anode plate 2 can also be formed by forming a thin film on the direct substrate 1 by electrolytic polymerization, or applying a conductive polymer on a substrate 1. The laminating of the different matter is carried out and an anode plate 2 can also form it.

[0022] The thickness of an anode plate 2 changes with existence of a demand of transparency. When transparency is needed, it is desirable to usually make the permeability of the light into 80% or more preferably 60% or more, and 5-1000nm of thickness is usually about 10-500nm preferably in this case. When opaque [an anode plate 2] and good, you may be the same ingredient as a substrate 1. Moreover, it is also possible to carry out the laminating of the different electrical conducting material on an anode

plate 2.

[0023] The organic luminous layer 3 formed on an anode plate 2 is formed in inter-electrode [to which electric field were given] from the ingredient which conveys efficiently the electron poured in from the electron hole and cathode 4 which were poured in from the anode plate 2, and is made to recombine it, and emits light efficiently by recombination. Usually, as this organic luminous layer 3 is shown in drawing 3 for improvement in luminous efficiency, making it the functional discrete type divided into electron hole transporting bed 3a and electron transport layer 3b is performed.

[0024] In functional discrete-type organic electroluminescence-devices 10A shown in drawing 3 , as an ingredient of electron hole transporting bed 3a, the hole-injection effectiveness from an anode plate 2 is high, and it is required to be the ingredient which can convey the poured-in electron hole efficiently. For that purpose, ionization potential is small, moreover, hole mobility is large, it excels in stability further, and it is required that it should be hard to generate the impurity which serves as a trap at the time of manufacture and an activity.

[0025] The aromatic series diamine compound which connected the 3rd class aromatic amine units, such as a 1 and 1-bis(4-G p-tolylamino phenyl) cyclohexane, as such an electron hole transport ingredient, for example, 4 and 4'-screw [N-(1-naphthyl)-N-phenylamino] Aromatic amine which two or more fused aromatic rings permuted by the nitrogen atom including two or more tertiary amine represented with a biphenyl, The aromatic series triamine which has starburst structure with the derivative of triphenyl benzene, Aromatic series diamines, such as N, N'-diphenyl-N, the N'-bis(3-methylphenyl) biphenyl -4, and 4'-diamine, alpha, alpha, alpha', alpha'-tetramethyl - alpha, alpha'-bis(4-G p-tolylamino phenyl)-para xylene, The compound which the aromatic series diamino radical permuted by the triphenylamine derivative unsymmetrical in three dimensions and the BIRENIRU radical as the whole molecule, The aromatic series diamine which connected the 3rd class aromatic amine unit by ethylene, The aromatic series diamine which has styryl structure, the thing which connected the aromatic series tertiary amine unit by the thiophene radical, Starburst mold aromatic series triamine, a benzylphenyl compound, the thing that connected tertiary amine by the fluorene radical, A triamine compound, a bis-pyridylamino biphenyl, N and N, N-triphenylamine derivative, The aromatic series diamine which has phenoxazine structure, a diamino phenyl phenanthridine derivative, a hydrazone compound, a silazane compound, a silanamine derivative, a phosphamine derivative, the Quinacridone compound, etc. are mentioned. These compounds may be used independently, and two or more sorts may be mixed and used if needed.

[0026] In addition, polymeric materials, such as a polyvinyl carbazole, polysilane, poly FOSUFAZEN, a polyamide, a polyvinyl triphenylamine, a giant molecule that has a triphenylamine frame, a giant molecule which connected the triphenylamine unit by the methylene group etc., and polymethacrylate containing aromatic amine, can be used as an ingredient of electron hole transporting bed 3a in addition to the above-mentioned compound.

[0027] Laminating formation of the electron hole transporting bed 3a is carried out on said anode plate 2 by forming these electron hole transport ingredients with vacuum evaporation technique, the sputtering method, electron beam vacuum deposition, etc.

[0028] In forming electron hole transporting bed 3a with a vacuum deposition method, after paying electron hole transport ingredients to the crucible installed in the vacuum housing and exhausting the inside of a vacuum housing to about 10 - 4Pa with a suitable vacuum pump, a crucible is heated, an electron hole transport ingredient is evaporated, and electron hole transporting bed 3a is formed on the anode plate 2 on the substrate 1 which carried out opposite arrangement at the crucible.

[0029] Thus, when forming electron hole transporting bed 3a, low-battery actuation can be enabled by doping the metal complex of aromatic carboxylic acid and/or a metal salt, a benzophenone derivative and a thio benzophenone derivative, and fullerene by 10⁻³ - 10% of the weight of concentration, and making the electron hole as a free carrier generate as an acceptor further.

[0030] 10-300nm of thickness of electron hole transporting bed 3a is usually 30-100nm preferably. In order to form uniformly the thin electron hole transporting bed of such thickness, generally a vacuum deposition method is used well.

[0031] Moreover, as shown in drawing 4 , forming hole injection layer 3c between electron hole

transporting bed 3a and an anode plate 2 is also performed, in order to raise hole-injection effectiveness further and to improve the adhesion force to the anode plate 2 of the whole organic layer. As an ingredient used for hole injection layer 3c, ionization potential is low, conductivity is high, the ingredient which can form a stable thin film thermally on an anode plate 2 further is desirable, and a phthalocyanine compound and a porphyrin compound are used. By making such hole injection layer 3c intervene, the effectiveness that the power surge when carrying out continuation actuation of the component by constant current is also controlled is acquired at the same time the driver voltage of an early component falls. It is possible to raise conductivity with hole injection layer 3c as well as electron hole transporting bed 3a doping an acceptor.

[0032] 2-100nm of thickness of hole injection layer 3c is usually 5-50nm preferably. In order to form the thin hole injection layer of such thickness uniformly, generally a vacuum deposition method is used well.

[0033] Electron transport layer 3b formed on electron hole transporting bed 3a consists of compounds which can convey the electron from cathode in the direction of electron hole transporting bed 3a efficiently in inter-electrode [to which electric field were given].

[0034] As an electronic transportability compound used for electron transport layer 3b, the electronic injection efficiency from cathode 4 is high, and it is required to be the compound which can convey the poured-in electron efficiently. For that purpose, an electron affinity is large, moreover electron mobility is large, it excels in stability further, and to be the compound which the impurity which serves as a trap at the time of manufacture and an activity cannot generate easily is demanded.

[0035] As an ingredient which fulfills such conditions, metal complexes, such as aromatic compounds, such as a tetra-phenyl butadiene, and an aluminum complex of 8-hydroxyquinoline, a cyclopentadiene derivative, a peri non derivative, an oxadiazole derivative, a bis-styryl benzene derivative, a perylene derivative, a coumarin compound, a rare earth complex, a JISUCHIRIRU pyrazine derivative, p-phenylene compound, a thiadiazolo pyridine derivative, a pyrrolo pyridine derivative, a NAFUCHI lysine derivative, etc. are mentioned.

[0036] Generally electron transport layer 3b using these compounds can play simultaneously the role which conveys an electron, and an electron hole and the role which brings about luminescence in the case of electronic recombination.

[0037] When electron hole transporting bed 3a has a luminescence function, electron transport layer 3b may play only the role which conveys an electron.

[0038] although being the object which changes the luminescent color, for example, doping fluorochromes for laser, such as a coumarin, by using the aluminum complex of 8-hydroxyquinoline as a host ingredient etc. is performed while raising the luminous efficiency of a component -- this invention -- also setting -- the above-mentioned organic electronic transportability ingredient -- a host ingredient -- carrying out -- various kinds of fluorochromes -- 10-3-10-mol % -- the luminescence property of a component can be further raised by doping.

[0039] 10-200nm of thickness of electron transport layer 3b is usually 30-100nm preferably.

[0040] Although electron transport layer 3b can also be formed by the same approach as electron hole transporting bed 3a, a vacuum deposition method is usually used.

[0041] In addition, as an organic luminous layer 3 of a monolayer mold which does not perform functional separation as shown in drawing 2, it is Pori (p-phenylenevinylene) and Pori which were mentioned previously. [2-methoxy-5-(2-ethylhexyloxy)-1 and 4-phenylenevinylene] Polymeric materials, such as Pori (3-alkyl thiophene), the system which mixed luminescent material and an electronic transition ingredient to macromolecules, such as a polyvinyl carbazole, are mentioned.

[0042] Cathode 4 plays the role which pours an electron into the organic luminous layer 3. Although the ingredient used as cathode 4 can use the ingredient used for said anode plate 2, in order to perform electron injection efficiently, its low metal of a work function is desirable, and suitable metals or those alloys, such as tin, magnesium, an indium, calcium, aluminum, and silver, are suitable for it. The thickness of cathode 4 is usually comparable as an anode plate 2.

[0043] When a work function carries out the laminating of the stable metal layer to atmospheric air

further highly on this cathode, the stability of a component can be increased in order to protect the cathode which consists of a low work function metal. Metals, such as aluminum, silver, nickel, chromium, gold, and platinum, are used for the metal layer for this object.

[0044] In addition, drawing 2 -4 can show an example of the component body adopted by this invention, and this invention can be applied to the component body of lamination as shown below in addition to the thing of a graphic display.

[0045] An anode plate / electron hole transporting bed / electron transport layer / volume phase / cathode, an electron transport layer/cathode besides anode plate / electron hole transporting bed / electron transport layer/, By an electron transport layer / the cathode above-mentioned lamination besides electron transport layer / volume phase / cathode, anode plate / hole injection layer / electron hole transporting bed / electron transport layer / volume phase / cathode, anode plate / hole injection layer / electron hole transporting bed / electron transport layer/[besides anode plate / electron hole transporting bed / electron transport layer/] A volume phase is for raising contact to cathode and an organic layer. An aromatic series diamine compound, The layer which consisted of the Quinacridone compound, a naphthacene derivative, the organic silicon compound, an organic phosphorous compound, a compound that has N-phenyl carbazole frame, an N-vinylcarbazole polymer, etc. can be illustrated. 2-100nm of thickness of a volume phase is usually 5-30nm preferably. Instead of preparing a volume phase, the field which contains the ingredient of the above-mentioned volume phase 50% of the weight or more near the cathode interface of an organic luminous layer and an electron transport layer may be prepared.

[0046] Moreover, in order that other electron transport layers may raise the luminous efficiency of organic electroluminescence devices further, it is required for the compound which laminating formation is further carried out on an electron transport layer, and is used for this electron transport layer that the electron injection from cathode should be easy and electronic transport capacity should be still larger. As such an electronic transportability ingredient, the system which distributed an oxadiazole derivative and them to resin, such as a polymethyl methacrylate (PMMA), a phenanthroline derivative or n mold hydrogenation amorphous carbonization silicon, n mold zinc sulfide, n mold zinc selenide, etc. are mentioned. 5-200nm of thickness of other electron transport layers is usually 10-100nm preferably.

[0047] In order to raise the stability and dependability of such organic electroluminescence devices, it is necessary to carry out the seal of the whole stratified deposit, such as the whole component, i.e., the anode plate formed on the substrate, an organic luminous layer, and cathode.

[0048] Below, sealing concerning this invention is explained with reference to drawing 5 . In addition, although the case where sealing of the protective layer 5 is prepared and carried out on the cathode 4 of organic electroluminescence-devices body 10B of the structure shown in drawing 4 is illustrated to drawing 5 , it cannot be overemphasized that you may be drawing 2 , the bodies 10 and 10A of organic electroluminescence devices shown in 3, or the organic electroluminescence-devices body of other lamination.

[0049] in drawing 5 , a protective layer 5 is formed in order to protect the cathode 4 and the organic layer under it -- having -- **** -- SiOx, and GeO, MgO and Ta 2O5 etc. -- an oxide and SiNx etc. -- sulfides, such as a nitride and GeS, and MgF2 etc. -- a fluoride etc. is used. As the formation approach, the vacuum evaporation technique by resistance heating, a spatter, electron beam vacuum deposition, etc. are used. Among these, although many vacuum evaporation technique is used in order to control the damage at the time of the protection stratification, using together the spatter formed into the low damage, and vacuum evaporation technique and a spatter is also performed. Generally thickness of a protective layer 5 is set to 100nm - 10 micrometers, and is suitably determined according to membranous precision, stress, and other properties and military requirements.

[0050] After forming a protective layer 5, except for the electrode section pulled out to particular parts, such as the circumference of a substrate 1, sealing is performed in order to intercept the component section (stratified deposit) from the external world. The seal member consists of a sealing compound (encapsulant) 6 and tooth-back glass 7 at least, after it applies a sealing compound 6 to a substrate 1 or tooth-back glass 7, it contacts both, it hardens a sealing compound 6, and completes sealing.

[0051] In this invention, it takes all over a dry box, without exposing the substrate 1 after forming a protective layer 5 to atmospheric air in this sealing, and this sealing is performed.

[0052] The thing of the space where the moisture content (water vapor content) occupied in environmental gas compared with a dry box here in usual atmospheric air was controlled low can be said, and inert gas, such as air, nitrogen, and an argon, and other handling matter and gas which does not react can be used as environmental gas. As an approach of making it into a dry environment, the gas dried [argon], for example permutes the inside of a box, and there is the approach of maintaining a dry environment by continuing passing small quantity [every] gas also after that. With this approach, it is [about / dew-point]. -10 degrees C (about 1600 ppm) are possible. Furthermore, in order to make it a low-water-flow daily dose, the equipment which carries out circulation purification of the environmental gas is installed separately, there is the approach of carrying out circulation operation, and it is possible according to this approach to maintain -100 degrees C (about 0.06 ppm) of below dew-point abbreviation.

[0053] Thermosetting resin, a photo-setting resin, etc. are used as a sealing compound 6. Also in which resin, compared with acrylic and a silicon system, epoxy system resin is excellent in respect of moisture permeability, and, generally is used well. The thickness of a sealing compound 6 needs to be thicker than the thickness of the sum total of the component (stratified deposit) formed in the substrate 1. For this reason, a silica bead, glass fiber, etc. may be used together with a sealing compound as a spacer in order to secure the thickness of the seal section.

[0054] Moreover, as a sealing agent on the back, although a metal, resin, etc. may be used other than tooth-back glass 7, also in which sealing agent, moisture permeability-proof, oxygen-proof permeability, etc. are required like a sealing compound. A sealing agent also takes into consideration properties, such as the visible permeability of a tooth-back sealing agent, reinforcement, and variability, and a selection activity is carried out again according to the application of a component.

[0055] Next, with reference to drawing 1, the manufacture approach of the organic electroluminescence devices by the manufacturing installation and this equipment of organic electroluminescence devices of this invention for manufacturing such organic electroluminescence devices is explained.

[0056] Drawing 1 is the mimetic diagram showing the gestalt of operation of the manufacturing installation of the organic electroluminescence devices of this invention.

[0057] They are the gate valve with which the vacuum chamber for conveyance, and 22b-26b connect a substrate and the robot arm for mask conveyance, and, as for a dry box, and 22-26, 22c-27c connect a dry box, the vacuum chamber for conveyance, or the vacuum chambers for conveyance for 21 and 27, as for an operating vacuum chamber, and 22a-26a, and the gate valve which connects an operating vacuum chamber and the vacuum chamber for conveyance 22d-26d among drawing 1.

[0058] After installing that by which the anode plate 2 by which patterning was carried out was formed on the substrate 1 in a dry box 21 in manufacture of organic electroluminescence devices and carrying out UV ozone washing of the substrate front face, inert gas fully permutes the inside of a dry box 21. Next, gate valve 22c is opened and a substrate is introduced in vacuum chamber 22a for conveyance using robot arm 22b. After closing gate valve 22c, vacuum suction of the vacuum chamber 22a for conveyance is carried out to 10 to 5 or less Toors, gate valve 22d is opened after that, a substrate is transported using robot arm 22b, and a substrate is installed in the operating vacuum chamber 22. In the operating vacuum chamber 22, plasma treatment of the substrate front face is carried out with the mixed gas of an argon and oxygen. Next, a substrate is transported through gate valve 22d and vacuum chamber 22 for conveyance a, gate valve 23c, vacuum chamber 23 for conveyance a, and gate valve 23d, using the robot arms 22b and 23b one by one, with a vacuum held, and a substrate is installed in the operating vacuum chamber 23. Within the operating vacuum chamber 23, sequential vacuum evaporation formation is carried out, and hole injection layer 3c and electron hole transporting bed 3a transport a substrate like the above after that, using the robot arms 23b and 24b one by one, and install a substrate in the operating vacuum chamber 24. Within the operating vacuum chamber 24, vacuum evaporation formation is carried out, and electron transport layer 3b transports a substrate like the above after that, using the robot arms 24b and 25b one by one, and installs a substrate in the operating

vacuum chamber 25. Within the operating vacuum chamber 25, vacuum evaporation formation of the cathode 4 is carried out, a substrate is transported like the above after that, using the robot arms 25b and 26b one by one, and a substrate is installed in the operating vacuum chamber 26. Within the operating vacuum chamber 26, vacuum evaporation formation is carried out, and a protective layer 5 installs a substrate in vacuum chamber 26a for conveyance by robot arm 26b after that, and is filled up with inert gas for gate valve 26d to atmospheric pressure in vacuum chamber 26a for closing conveyance.

[0059] Next, gate valve 27c is opened, in the dry box 27 beforehand filled with inert gas, robot arm 26b is used and a substrate is installed. In a dry box 27, after applying the photo-setting resin of a sealing compound 6 on a substrate 1, tooth-back glass 7 is made to contact and the seal of the organic electroluminescence-devices body 10B is carried out.

[0060] As mentioned above, according to this equipment, a substrate is not exposed to atmospheric air after filling up with the inert gas after UV ozone washing in a dry box 21 until a seal is completed in a dry box 27.

[0061] Thus, it is taken out from a dry box 27, the electric wiring for component actuation and mounting of a semiconductor circuit are made, and the organic electroluminescence devices which the seal completed are completed as a panel.

[0062] In addition, although the unit for migration and processing which consists of an operating vacuum chamber, a vacuum chamber for conveyance, etc. is connected with the five-piece serial in the shape of a straight line in drawing 1. This number of connection units does not necessarily need to be five pieces, and it can also perform easily being able to connect the number of two or more arbitration by the class of the number of stratified deposits, or stratified deposit, the other arts of a substrate, etc., and fluctuating the number of units according to the change in a stratified deposit, modification of an art, etc. Moreover, two dry boxes are not necessarily required, either and it is also possible to collect two functions to one or to omit one function. A robot arm needs a certain thing for one or more vacuum chambers for conveyance, and when two or more [of these] are prepared, migration of a substrate and a mask becomes efficient and it can shorten working hours more.

[0063] A robot arm here points out the thing of the device which holds a substrate and a mask and can move between vacuum chambers, the maintenance means or migration means of the configuration of equipment and a substrate is not specified at all, and combination with a conveyor type migration means etc. is also possible.

[0064] Moreover, the processing in the operating vacuum chambers 22-26 is not limited to the approach explained above at all, but can adopt various arts, such as a spatter and an electron beam method, as the membrane formation approach. Moreover, ultraviolet rays and an excimer laser can be irradiated in addition to the plasma treatment method according to the mixed gas of argon gas and oxygen gas also as an art of the substrate in the operating vacuum chamber 22, or various substrate arts, such as the approach of exposing to specific gas and reforming a substrate front face, can be adopted. For example, in the operating vacuum chamber 22, the plasma or laser radiation performs refining of a substrate, in the operating vacuum chambers 23-25, with the vacuum deposition by resistance heating, each organic layer and cathode can be formed and a protective layer can also be formed by the spatter in the operating vacuum chamber 26. Moreover, in one operating vacuum chamber, it can also have two or more approaches, each approach is used, and the membrane formation approach and a substrate art can also be formed membranes or processed sequential or simultaneous. Moreover, although the art after carrying out pattern formation of the anode plate as a lower electrode here was indicated, it is also possible to also form a lower electrode in the equipment of a graphic display. Moreover, the order of formation of an anode plate and cathode was not fixed, either, and it can choose freely to compensate for the configuration of an organic layer.

[0065] This invention is applicable also to a single component, the component which consists of structure arranged in the shape of an array, and which organic electroluminescence devices of the structure where an anode plate and cathode have been arranged in the shape of an X-Y matrix.

[0066]

[Example] Next, although an example and the example of a comparison are given and this invention is

explained still more concretely, this invention is not limited to the publication of the following examples, unless the summary is exceeded.

[0067] Using 7059 by Corning, Inc. glass with a thickness of 1.1mm as example 1 glass substrate, 120nm deposition (Geomatec electron beam membrane formation article; 20ohms of sheet resistance) of the indium stannic-acid ghost (ITO) transparence electric conduction film was carried out on it, and the glass substrate with the ITO film was obtained.

[0068] Next, the organic electroluminescence devices which have the structure shown in drawing 5 using the equipment shown in drawing 1 were produced by the following approaches.

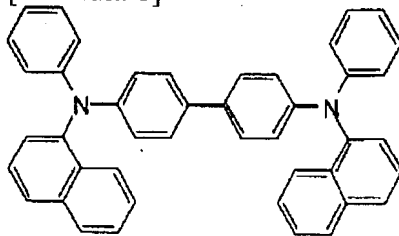
[0069] Patterning of the ITO transparence electric conduction film deposited on the glass substrate 1 was carried out to the stripe of 2mm width of face using the usual photolithography technique and hydrochloric-acid etching, and the anode plate 2 was formed. It was made to dry by nitrogen blow after washing in order of ultrasonic cleaning by the acetone, rinsing by pure water, and ultrasonic cleaning by isopropyl alcohol, and the ITO substrate which carried out pattern formation was installed in the dry box 21. In the dry box 21, after performing UV / ozone washing for 10 minutes, inert gas replacement of the inside of a dry box was carried out with nitrogen gas, gate valve 22c was opened, and the substrate was introduced in vacuum chamber 22a for conveyance by robot arm 22b. After closing gate valve 22c, vacuum suction of the inside of vacuum chamber 22a for conveyance was carried out also using the cryopump to 1.1×10^{-6} Torr (about 1.5×10^{-5} to 4×10^{-5} Pa). Then, gate valve 22d was opened and the substrate 1 was installed in the operating vacuum chamber 22 using robot arm 22b. After closing gate valve 22d, in the operating vacuum chamber 22 which was the vacua of 1.0×10^{-6} Torr, mixed gas of 50% of oxygen was introduced argon 50%, until it was set to 1.0×10^{-3} Torr, and the front face of the glass substrate 1 with ITO was processed for 5 minutes in the plasma. Installation of the blasting fumes was suspended, and vacuum suction of the inside of the operating vacuum chamber 22 was carried out until it would be in the vacua of 1.0×10^{-6} Torr again.

[0070] Next, gate valve 22d was opened, the substrate 1 was transported into vacuum chamber 22a for conveyance from the operating vacuum chamber 22 using robot arm 22b, after closing gate valve 22d, gate valve 23c was opened, the substrate 1 was transported into vacuum chamber 23a for conveyance using robot arm 22b and robot arm 23b, and gate valve 23c was closed. While the substrate 1 was transported to 23a from vacuum chamber 22 for conveyance a, the inside of vacuum chamber 22 for conveyance a and 23a was maintained at the vacua of 1.0×10^{-6} Torr.

[0071] Next, gate valve 23d was opened and the substrate 1 was installed in the operating vacuum chamber 23 using robot arm 23b. Under the present circumstances, a substrate 1 is installed on the metal mask arranged beforehand, and the part for the anode plate ejection on a substrate 1 was covered with the metal mask. Next, it vapor-deposited by heating the copper phthalocyanine (H1) (crystal form being beta mold) which shows gate valve 23d to the following put into the molybdenum boat arranged in closing and the operating vacuum chamber 23. It vapor-deposited in degree of vacuum 1.1×10^{-6} Torr (about 1.5×10^{-5} to 4×10^{-5} Pa), and vacuum evaporation time amount 1 minute, and hole injection layer of 20nm of thickness 3c was obtained.

[0072]

[Formula 1]



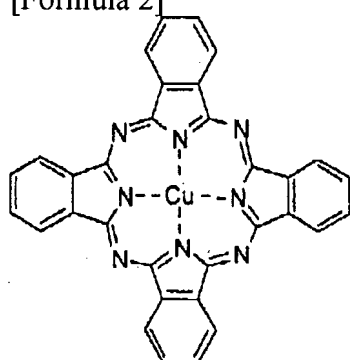
(H 1)

[0073] Next, 4 and 4' bis[-] [N-(1-naphthyl)-N-phenylamino] biphenyl (H2) shown in the following put into the ceramic crucible arranged in the operating vacuum chamber 23 was heated at the tantalum wire heater around a crucible, and the laminating was carried out on hole injection layer 3c. The temperature

of the end crater which gets at this time was controlled in 230-240 degrees C. Electron hole transporting bed of 60nm of thickness 3a was obtained in degree of vacuum 8×10^{-7} Torr at the time of vacuum evaporation (about 1.1×10 to 4 Pa), and vacuum evaporation time amount 50 seconds per minute.

[0074]

[Formula 2]

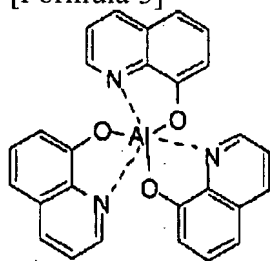


(H 2)

[0075] Next, gate valve 23d was opened, the substrate 1 was transported into vacuum chamber 23a for conveyance from the operating vacuum chamber 23 using robot arm 23b, after closing gate valve 23d, gate valve 24c was opened, the substrate 1 was transported into vacuum chamber 24a for conveyance using robot arm 23b and robot arm 24b, and gate valve 24c was closed. Then, gate valve 24d was opened and the substrate 1 was installed in the operating vacuum chamber 24 using robot arm 24b. Under the present circumstances, a substrate 1 is installed on the metal mask arranged beforehand, and the part for the anode plate ejection on a substrate 1 was covered with the metal mask. Next, 8-hydroxyquinoline complex $\text{Al}(\text{C}_9\text{H}_6\text{NO})_3$ (E1) of the aluminum which shows gate valve 24d with the following structure expressions as an ingredient of electron transport layer 3b which has a luminescence function within closing and the operating vacuum chamber 24 was vapor-deposited similarly on the above-mentioned electron hole transporting bed 3a. The temperature of the end crater which gets at this time was controlled in 310-320 degrees C. The degree of vacuum at the time of vacuum evaporation was 9×10^{-7} Torr (about 1.2×10 to 4 Pa), vacuum evaporation time amount was 2 minutes and 40 seconds, and the thickness of vapor-deposited electron transport layer 3b was 75nm.

[0076]

[Formula 3]



(E 1)

[0077] In addition, the substrate temperature when carrying out vacuum deposition of hole injection layer 3c, above-mentioned electron hole transporting bed 3a, and above-mentioned electron transport layer 3b was held to the room temperature.

[0078] Next, gate valve 24d was opened, the substrate 1 was transported into vacuum chamber 24a for conveyance from the operating vacuum chamber 24 using robot arm 24b, after closing gate valve 24d, gate valve 25c was opened, the substrate 1 was transported into vacuum chamber 25a for conveyance using robot arm 24b and robot arm 25b, and gate valve 25c was closed. Gate valve 25d was opened after that, and the substrate 1 was installed in the operating vacuum chamber 25 using robot arm 25b. Under the present circumstances, a substrate 1 is installed on the metal mask for cathode vacuum evaporation arranged beforehand, and the stripe-like shadow mask of 2mm width of face was stuck to the substrate 1 so that it might intersect perpendicularly with the ITO stripe of the patternized anode plate 2.

[0079] Next, within closing and the operating vacuum chamber 25, as cathode 4, gate valve 25d was vapor-deposited so that 2 μm might become 100nm of thickness with simultaneous vacuum deposition about the alloy electrode of magnesium and silver. Vacuum evaporation was performed using the molybdenum boat in degree of vacuum 1×10^{-5} Torr (about 1.3×10^{-3} Pa), and vacuum evaporation time amount 3 minutes and 10 seconds. Moreover, the atomic ratio of magnesium and silver was set to 10:1.2. Furthermore it continued, the laminating of the aluminum was carried out on magnesium and the silver alloy film by 100nm thickness using the molybdenum boat into the operating vacuum chamber 25, and cathode 4 was completed. The degree of vacuum at the time of the vacuum plating of aluminium was 2.3×10^{-5} Torr (about 3.1×10^{-3} Pa), and vacuum evaporation time amount was 40 seconds per minute. The substrate temperature at the time of vacuum evaporation of the above magnesium and silver alloy, and the two-layer mold cathode of aluminum was held to the room temperature.

[0080] Next, gate valve 25d was opened, the substrate 1 was transported into vacuum chamber 25a for conveyance from the operating vacuum chamber 25 using robot arm 25b, after closing gate valve 25d, gate valve 26c was opened, the substrate 1 was transported into vacuum chamber 26a for conveyance using robot arm 25b and robot arm 26b, and gate valve 26c was closed. Gate valve 26d was opened after that, and the substrate 1 was installed in the operating vacuum chamber 26 using robot arm 26b. Under the present circumstances, the substrate 1 was stuck and installed on the metal mask for protective layer vacuum evaporation arranged beforehand. Next, within closing and the operating vacuum chamber 26, as a protective layer 5, gate valve 26d was vapor-deposited so that it might become 1000nm of thickness about silicon oxide ($\text{SiO}_x; x=1.0-2.0$). Vacuum evaporation was performed using the molybdenum boat in degree of vacuum 1×10^{-5} Torr (about 1.3×10^{-3} Pa), and vacuum evaporation time amount 3 minutes and 40 seconds. The substrate temperature at the time of this vacuum evaporation was held to the room temperature. Thereby, the component section except the ejection parts of an anode plate and cathode was covered by the protective layer 5.

[0081] Next, after having opened gate valve 26d, transporting the substrate 1 into vacuum chamber 26a for conveyance from the operating vacuum chamber 26 using robot arm 26b and closing gate valve 26d, nitrogen was introduced in vacuum chamber 26a for conveyance, and it considered as atmospheric pressure.

[0082] Then, gate valve 27c was opened, in the dry box 27 currently beforehand filled with nitrogen gas, robot arm 26b was used, and the substrate 1 was installed, next dispensing of the photo-setting resin (30Y-184) by Three Bond Co., Ltd. was carried out to the seal section on the substrate 1 of organic electroluminescence-devices body 10B by width of face of 0.5mm as a sealing compound 6. Then, they are 50 g/cm² so that a sealing compound 6 may install the tooth-back glass 7 beforehand cut into the size of the seal section in the part carried out with ** and a glass substrate 1 and tooth-back glass 6 may stick it. The pressure was put for 30 seconds.

[0083] Next, it is ultraviolet radiation (wavelength of 365nm) to the part to which the sealing compound 6 was carried out with ** 4.2 J/cm² It irradiated, the photo-setting resin was stiffened and the seal was made to complete. On the occasion of hardening, in order to prevent degradation of the component by ultraviolet rays and heat, parts other than the seal section shaded.

[0084] Then, ejection and the organic electroluminescence devices of 2mmx2mm size were obtained for the component from the ejection room established in the dry box 27.

[0085] Impress the direct current voltage of minus, it was made to emit light to the anode plate 2 of the obtained organic electroluminescence devices in plus and cathode 4, and the luminescence property was measured. Current density 15 mA/cm² The time amount change to the initial value of the luminescence brightness when continuing passing a current for a component is shown in drawing 6.

[0086] The luminescence brightness in early stages of [energization] the organic electroluminescence devices by this example was 9250 cd/m². The luminescence brightness after 10000-hour progress is 8650 cd/m². It became, the relative luminance to initial value was 0.935, and lowering of brightness was small.

[0087] The organic electroluminescence devices of the same lamination as the organic electroluminescence devices manufactured in the example 1 for the example of comparison 1

comparison were produced by the option.

[0088] That is, except for having once taken out the rear spring supporter and the substrate 1 from the operating vacuum chamber to 2 times after formation of a protective layer 5 into atmospheric air the formation back of electron transport layer 3b, and having moved to the following process using the isolated system, others produced organic electroluminescence devices on the same conditions as an example 1, without two or more operating vacuum chambers connecting.

[0089] The configuration, the construction material, the thickness, and the appearance of a layer of the obtained organic electroluminescence devices were the same as that of the thing of an example 1.

[0090] About these organic electroluminescence devices, luminescence brightness was measured by the same approach as an example 1, and the time amount change to the initial value of luminescence brightness was shown in drawing 6.

[0091] the organic electroluminescence devices of this example of a comparison -- the luminescence brightness in early stages of energization -- 9100 cds/m² it is -- there were not a thing of an example 1 and a big difference. However, the luminescence brightness after 1000-hour progress is 5105 cd/m². The relative luminance to initial value is set to 0.561, and 10000 hours after, it will not shine, so that measurement of luminescence brightness is impossible.

[0092] The organic electroluminescence devices of the same lamination as the organic electroluminescence devices manufactured in the example 1 for the example of comparison 2 comparison were further produced by the option.

[0093] That is, except for having once taken out the substrate 1 from the operating vacuum chamber into atmospheric air after formation of a protective layer 5, and having moved to the following seal process using the equipment with which two or more operating vacuum chambers were connected with the perimeter of one vacuum chamber for conveyance, others produced organic electroluminescence devices on the same conditions as an example 1.

[0094] The configuration, the construction material, the thickness, and the appearance of a layer of the obtained organic electroluminescence devices were the same as that of the thing of an example 1.

[0095] About organic electroluminescence devices, luminescence brightness was measured by the same approach as an example 1, and the time amount change to the initial value of luminescence brightness was shown in drawing 6.

[0096] the organic electroluminescence devices of this example of a comparison -- the luminescence brightness in early stages of energization -- 9150 cds/m² it is -- there were not a thing of an example 1 and a big difference. However, the luminescence brightness after 10000-hour progress is 6680 cd/m². Although the relative luminance to initial value was set to 0.73 and its lowering of brightness was smaller than the example 1 of a comparison, compared with the example 1, its lowering of brightness was large.

[0097]

[Effect of the Invention] According to this invention, each class-like deposit prevents contacting the moisture and oxygen in atmospheric air by carrying out without being consistent also including each process and its shift process from after the pattern formation of an anode plate to sealing, and exposing a substrate to atmospheric air in manufacture of organic electroluminescence devices, productivity is good and organic electroluminescence devices excellent in the luminescence property and the luminescence life property can be offered by low cost as explained in full detail above.

[0098] The application to the light source (for example, the light source of a copying machine, the back light light source of a performance display or instruments) and the plotting board with which the organic electroluminescence devices manufactured by this invention employed the description as a flat-panel display (for example, the object for OA computers and a flat TV) or a field illuminant efficiently, and a beacon light is expected, and the technical value is very large.

[Translation done.]

*** NOTICES ***

JPO and INPIT are not responsible for any damages caused by the use of this translation.

- 1.This document has been translated by computer. So the translation may not reflect the original precisely.
- 2.**** shows the word which can not be translated.
- 3.In the drawings, any words are not translated.

DESCRIPTION OF DRAWINGS

[Brief Description of the Drawings]

[Drawing 1] It is the mimetic diagram showing the gestalt of operation of the manufacturing installation of the organic electroluminescence devices of this invention.

[Drawing 2] It is the typical sectional view showing one example of the organic electroluminescence-devices body manufactured by this invention.

[Drawing 3] It is the typical sectional view showing other examples of the organic electroluminescence-devices body manufactured by this invention.

[Drawing 4] It is the typical sectional view showing another example of the organic electroluminescence-devices body manufactured by this invention.

[Drawing 5] It is the typical sectional view showing the example of the organic electroluminescence devices manufactured by this invention.

[Drawing 6] It is the graph which shows the actuation luminescence time dependency of the relative luminescence brightness to the initial luminescence brightness of the organic electroluminescence devices in an example 1 and the examples 1 and 2 of a comparison.

[Description of Notations]

- 1 Substrate
- 2 Anode Plate
- 3 Organic Luminous Layer
- 3a Electron hole transporting bed
- 3b Electron transport layer
- 3c Hole injection layer
- 4 Cathode
- 5 Protective Layer
- 6 Sealing Compound
- 7 Tooth-Back Glass
- 10, 10A, 10B Organic electroluminescence-devices body
- 21 27 Dry box
- 22, 23, 24, 25, 26 Operating vacuum chamber
- 22a, 23a, 24a, 25a, 26a Vacuum chamber for conveyance
- 22b, 23b, 24b, 25b, 26b Robot arm
- 22c, 23c, 24c, 25c, 26c, 27c Gate valve
- 22d, 23d, 24d, 25d, 26d Gate valve

[Translation done.]

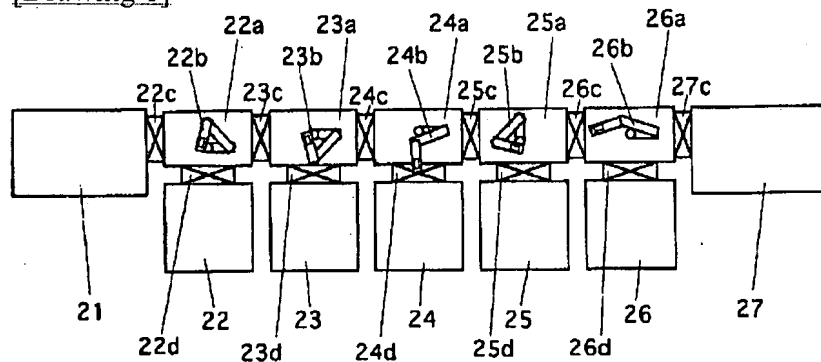
* NOTICES *

JPO and INPIT are not responsible for any damages caused by the use of this translation.

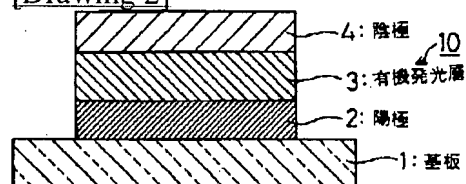
- 1.This document has been translated by computer. So the translation may not reflect the original precisely.
- 2.**** shows the word which can not be translated.
- 3.In the drawings, any words are not translated.

DRAWINGS

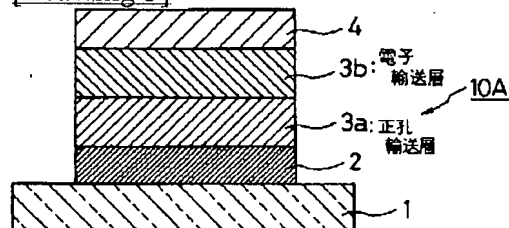
[Drawing 1]



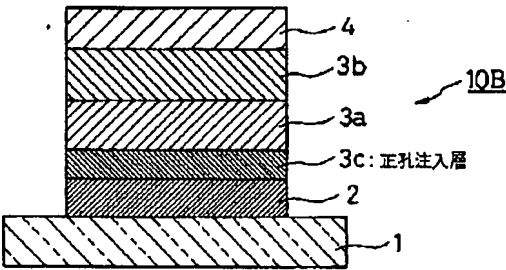
[Drawing 2]



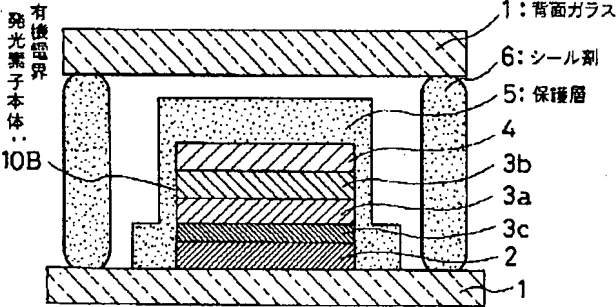
[Drawing 3]



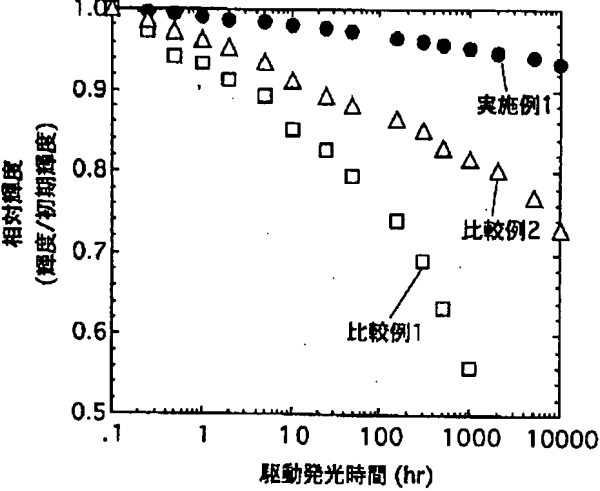
[Drawing 4]



[Drawing 5]



[Drawing 6]



[Translation done.]